

Confinement Induced Quantum Phase Transition and Polarization Cooling in a Dipolar Crystal of Polar Molecules

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It is well-known that the liquid properties in a strongly confined system can be very different from their ordinary behaviors in an extended system, due to the competition between the thermal energy and the interaction energy. Here we show that, in a low-dimensional self-assembled dipolar crystal, the parabolic optical confinement potential can also strongly affect the quantum many-body properties in the low temperature regime. For example, by changing the confinement aspect ratio, the bulk of the system can undergo a quantum phase transition between a liquid state and a solid state via a nonmonotonic pattern formation of the domain wall. Furthermore, the entropy of a trapped dipolar crystal can be much larger than the liquid state in the weak dipole limit, indicating an intrinsic polarization cooling mechanism via increasing the external field. These highly correlated confinement effects are very important to the experimental preparation of a self-assembled dipolar crystal using ultracold polar molecules.

Confinement effects are known to be very crucial to the physical properties of a spatially confined liquid. Investigating these extraordinary fluid properties has become an important subject in nanoscience and technology in recent years [1]. When the confined length scale is reduced to be comparable to the inter-particle distance, there could be several kinds of phase transitions observed in different systems, including solidification on the boundary [2], liquid-to-liquid phase transition [3], and layering phenomena [4], etc. From a microscopic point of view, all these unusual properties result from the competition between the thermal kinetic energy and the inter-particle interaction in a certain spatial geometry. It is therefore reasonable to expect that the interplay between quantum fluctuation effects, interaction energy, and the confinement geometry can also lead to some extraordinary many-body properties that are not observable in classical systems. Here we show that in a self-assembled low-dimensional dipolar crystal [5], which can be formed by ultracold polar molecules trapped in a magneto-optical potential, the bulk properties of the crystal can be strongly affected by the confinement potential, even when the system size is still much larger than the average inter-particle distance. The thermodynamical properties are also found qualitatively different from their behavior in an extensive space, showing the highly correlated confinement effect of a quantum many-body system. Different from the observed dipolar and confinement effects in the condensate of magnetic atoms [6], here we mainly focus on the dipolar crystal phase in the strongly interacting regime (see below), which cannot be easily achieved by magnetic atoms within present experimental parameter range.

We first consider polar molecules initially prepared in the lowest rotational ground state ($L = 0$) and confined in a quasi-two-dimensional (2D) trap, where the transverse dynamics (in z axis) is frozen to the single particle ground state. The in-plane motion of molecules, however, is weakly confined by weaker harmonic potentials with

trapping frequencies, $\omega_{x,y}$, in the x and y directions respectively. When an external DC electric field is applied along the z axis, polar molecules become polarized and have a field-dependent electric dipole moment, D . In a dilute limit, the mutual interaction between these quasi-2D polar molecules is a dipolar interaction [7], and the system can be easily stabilized by the strong transverse confinement potential. As a result, the system Hamiltonian can be written to be

$$H = \sum_i^N \left[\frac{\mathbf{p}_i^2}{2m} + \frac{m}{2} (\omega_x^2 x_i^2 + \omega_y^2 y_i^2) \right] + \frac{1}{2} \sum_{i \neq j}^N \frac{D^2}{|\mathbf{r}_i - \mathbf{r}_j|^3}, \quad (1)$$

where m is the mass and N is the total number of molecules. \mathbf{p}_i and $\mathbf{r}_i = (x_i, y_i)$ are the in-plane momentum and position operators of the i th particles. There are three length scales in Eq. (1): two are the oscillator lengths, $a_{\text{osc},x(y)} \equiv \sqrt{\hbar/m\omega_{x(y)}}$, and one is the dipolar interaction strength, $a_d \equiv mD^2/\hbar^2$. The system properties are therefore determined by three dimensionless parameters only: $\gamma \equiv a_d/a_{\text{osc},x}$, measuring the dipolar interaction strength, $\kappa \equiv \omega_y/\omega_x = a_{\text{osc},x}^2/a_{\text{osc},y}^2$, measuring the aspect ratio of the confinement potential, and particle number N . Throughout this paper, we use $a_{\text{osc}} \equiv a_{\text{osc},x}$ and $\omega \equiv \omega_x$ to be the units of length and energy scales respectively. For a typical molecule like SrO, the full polarized dipole moment is $D = 8.9$ Debye and therefore $\gamma \sim 88$ for $a_{\text{osc}} = 1.4\mu\text{m}$ in a typical trapping frequency, $\omega = 2\pi \times 50$ Hz.

In this paper, we are interested in the regime when the dipolar interaction is sufficiently large ($\gamma \gg 1$), so that polar molecules can form a dipolar crystal [5], as shown in Fig. 1(a) (for 1D trap, $\kappa = \infty$) and Fig. 1(e) (for 2D isotropic trap, $\kappa = 1$). Here the equilibrium position for each molecule is calculated by using Molecular-Dynamics(MD) simulation with a small friction in the Langevin equation [8,9]. We further calculate the phonon spectrum by quantizing the position fluctuation of dipoles from their equilibrium positions to the

quadratic order. Within this harmonic approximation, the obtained spectrum (Figs. 1(b) and (f)) is independent of the dipolar strength (γ), because the effect of tuning dipole moment can be exactly cancelled by adjusting the inter-molecule distance [10]. In Fig. 1(c)-(d) and (g)-(h), we show some typical phonon excitation wavefunctions for these two systems respectively. One easily see that the confinement potential has strongly changed the excitation wavefunctions from simple plane waves in a uniform system.

Following the same spirit of the Lindermann criterion, which was first applied to the classical 3D melting problem [12] and then extended to the 2D thermal and quantum melting [13,14,15], in this paper we define that a solid phase is melted at a certain position if its position fluctuation is larger than an imperial ratio (C_L) of the lattice constant. Such ratio in a uniform 2D dipolar crystal has been calculated to be 0.23 from the quantum Monte Carlo simulation [14,16]. To apply such useful criterion to a nonuniform system discussed in this paper, here we define the local “softness” of the dipolar crystal to be: $\xi_i \equiv \Delta r_i / \bar{l}_i$, where $\Delta r_i \equiv \sqrt{\langle (\mathbf{r}_i - \mathbf{r}_{i,0})^2 \rangle}$ is the position fluctuation of the i th particle (calculated from the wavefunction of the phonon excitation states), and \bar{l}_i is its average distance to the nearest neighboring sites. As a result, the i th particle is considered to be in a liquid state if $\xi_i > C_L$, while it is in a solid state if $\xi_i < C_L$ [18]. In Fig. 2(a) and (b), we show the local softness (ξ_i) for dipolar crystal in both 1D and 2D isotropic potentials. We find that in 1D system the softness distribution is convex with its maximum value in center of trap, i.e. the bulk of the 1D dipolar crystal is softer than the edge, in contradiction to a naive guess from a local density approximation, which predicts the position of higher particle density should be more solid-like due to the r^{-3} dipolar interaction. This striking results originate from the confinement effect on the quantum fluctuation of lattice points: the system ground state wavefunction has equal contribution from each eigenmodes (note that quantum zero point energy is $E_Q = \sum_n \frac{1}{2} \hbar \omega_n$), while higher energy modes prefer to generate much larger position fluctuation in the center of the 1D trap (see Fig. 1 (c) and (d)). In the 2D isotropic trap, on the other hand, the softness becomes concave, because now the high energy eigenmodes can still have large amplitude at the edge due to the additional degree of freedom in the azimuthal direction of the trap (see Fig. 1(g) and (h)). In Fig. 2(c) and (d), we further show the phase boundary (domain wall) between the liquid state and the solid(crystal) state as a function of dipolar strength (γ): the 1D system has a central domain of liquid state as long as γ is smaller than a critical value, γ_{1D}^* , above which the whole system becomes a well-defined solid crystal. On the other hand, the center of the 2D isotropic trap starts to be crystalized only when γ is larger than another critical value, γ_{2D}^* . Such qualitative difference in these two systems are clear evidence of the highly correlated confinement effects on the quantum fluctuations. Both γ_{1D}^* and γ_{2D}^* decrease as the

number of particles increases.

In Fig. 3, we show the confinement effects in different trap aspect ratio with a fixed ω_x : the anisotropic trapping potential generates two local minimum of the softness (see Fig. 3(b)) along the x axis. Within a proper parameter range, these two local minimum of softness can become the centers of two solid island(domain) embedded in a dipolar liquid. In Fig. 3(c), we show how the local softness, in the edge (red lines) and in the center (black lines) of the trap, changes as function of the aspect ratio, κ . We find that although the former decreases monotonically as expected, the later can have several re-entrant effects and can be even larger (i.e. softer) than its value in the isotropic trap ($\kappa = 1$) without compression. Increasing number of particles does not change these nonmonotonic bulk properties (say domain walls and re-entrant behavior), showing a significant confinement effects even when the system size is much larger than inter-particle distance. Similar nonmonotonic/reentrant behavior are also observed in a system of classical 2D melting [15] but much more particles and quantum fluctuation effects are considered here.

The nontrivial confinement effect can be also observed in the finite(but low) temperature regime by investigating the system entropy (S), which can be assumed to be conserved during an adiabatic manipulation of the system parameters. The total entropy can be calculated from [19] $S = \int_0^T dT' C_v(T')/T'$, where $C_v(T) = \partial E(T)/\partial T$ is the specific heat. The total energy, $E(T)$, can be easily calculated from the phonon excitation spectrum: $E(T) = E_C + E_Q + \sum_n \frac{\hbar \omega_n}{e^{\hbar \omega_n / k_B T} - 1}$, where E_C is classical potential energy and k_B is Boltzman constant. In Fig. 3(d), we show how the system temperature changes as a function of the confinement aspect ratio, κ , by keeping the total entropy a constant during the adiabatic process: the temperature increases sublinearly as the system is compressed and eventually becomes saturated in the limit of pure 1D system (say $\kappa > 100$ for $N = 91$). Within the harmonic expansion approximation for the phonon excitations, the calculate system entropy is independent of the dipolar strength, γ .

From experimental point of view, one of the most important question is how the system temperature changes when a dipolar crystal is formed by increasing the electric field. In the zero dipole moment limit (i.e. zero external field), only s -wave scattering exists between bosonic molecules, while it is almost noninteracting for single component fermionic molecules. For a Bose liquid in a 2D isotropic trap, we can apply the local density approximation to calculate the system entropy [20], and obtain the leading order temperature dependence: $S_{BL}/k_B = \alpha \frac{\pi^2}{\sqrt{2}} \left(\frac{k_B T}{\hbar \omega} \right)^2$ with $\alpha \sim 5.7 \times 10^{-3}$ being obtained from direct numerical calculation. This result is independent of the s -wave scattering length and particle number due to the unique geometry of 2D isotropic harmonic trap. Similar to the 3D case [20], it applies to condensate as well as to the normal state, because the

dominate contribution of entropy always comes from the single particle excitation of normal liquid. For the noninteracting fermionic molecules, we can also calculate the system entropy easily from the temperature dependence of the total energy in a 2D harmonic trap [21], and obtain $S_{\text{FL}}/k_B = \frac{\pi^2 \sqrt{2N}}{3} \left(\frac{k_B T}{\hbar \omega} \right)$. In Fig. 4 we show the calculated entropies of Fermi liquid (dotted lines), Bose liquid (dash-dotted line) and dipolar lattice (solid lines) in a 2D isotropic trap. Results for both $N = 91$ and $N = 217$ are shown together for comparison. It is easy to see that if bosonic polar molecules are initially prepared at zero field and in a sufficiently low temperature ($< T_{\text{boson}}^*$), the system temperature will decrease (intrinsic cooling, blue leftward arrow) as the external field is increased to derive the system toward a dipolar crystal adiabatically. On the other hand, if the system is prepared in a rather high initial temperature ($> T_{\text{boson}}^*$), the system temperature will increase greatly as the dipole moment increases (intrinsic heating, the red rightward arrow). The critical temperature, $T_{\text{boson}}^* \sim 10\hbar\omega/k_B$ for $N = 91$, but becomes about $16\hbar\omega/k_B$ for $N = 217$. This shows that the polarization cooling here is due to the many-body effects of polar molecules, completely different from the demagnetization cooling process in solid state systems or in magnetic dipolar atoms [22]. More precisely, due to the presence of a harmonic confinement, the average dis-

tance between dipoles increases (i.e. the average density decreases) as the dipole moment is enhanced, and therefore the system temperature can be reduced via transferring the electric field energy to the confinement potential energy (rather than to the kinetic energy) in the limit of strong dipolar crystal. Such intrinsic cooling mechanism is totally different from what is expected in a uniform system, where the entropy of a crystal should be always smaller than a liquid state at the same density [23]. Similar intrinsic cooling mechanism can be also observed in fermionic polar molecules and/or in 1D trapped system.

In summary, we have shown several important confinement effects on a self-assembled dipolar lattice formed by ultracold polar molecules. Changing confinement aspect ratio can induce a quantum phase transition between a bulk liquid state in 1D trap to a bulk solid crystal state in 2D isotropic trap. We further find an intrinsic polarization cooling mechanism during the formation of dipolar crystal. Our results can be applied to the experimental preparation of a dipolar crystal of ultracold polar molecules and are also important to the understanding of the confinement effects in a quantum many-body system.

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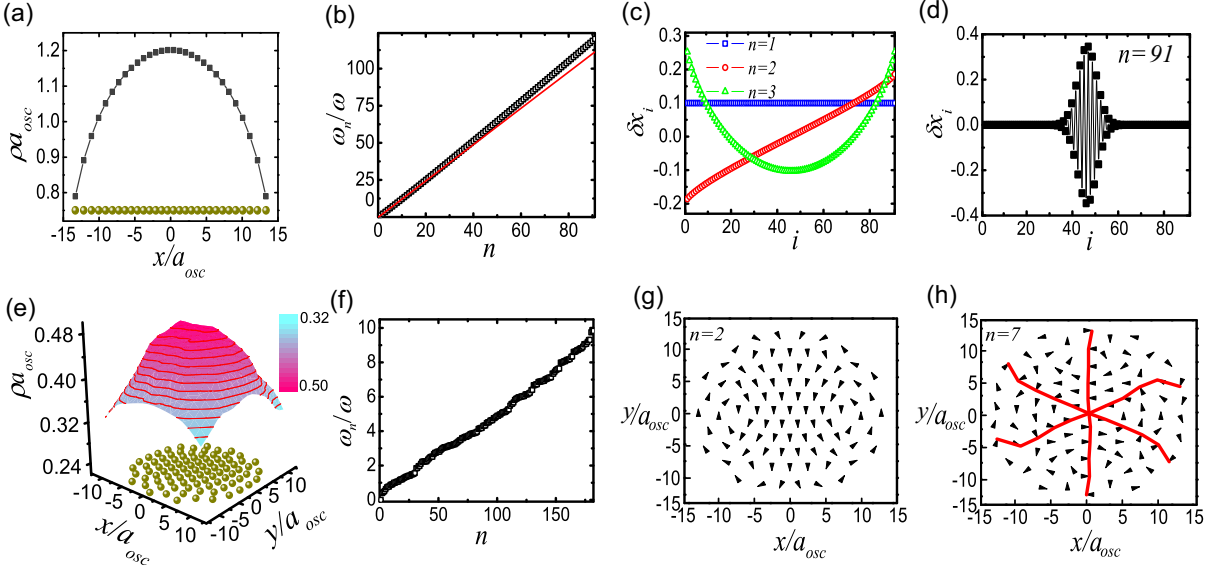


FIG. 1: (a) Density profile and position configuration of lattice position for $N = 30$ dipolar particles in a 1D harmonic trap with $\gamma = 17$. (b) Excitation spectrum for the same system with $N = 91$. The red line is obtained in the hydrodynamic theory of the liquid state with $N \rightarrow \infty$: $\omega_n = \omega_x \sqrt{(3n^2 - n)/2}$ [10]. (c) and (d) show the eigenfunction (i.e. deviation from their equilibrium position) for the lowest ($n = 1, 2, 3$) and the highest ($n = 91$) excited states of 1D system. (e)-(h) show the same physical quantities as (a)-(d), but for $N = 91$ particles in a 2D isotropic harmonic trap with $\gamma = 80$. (g) and (h) are the second and the seventh excitation modes with vibrating direction shown by arrows. The red lines in (h) are eye-guiding, indicating a Tachenko's mode as observed in the vortex lattice of a fast rotating condensate [11].

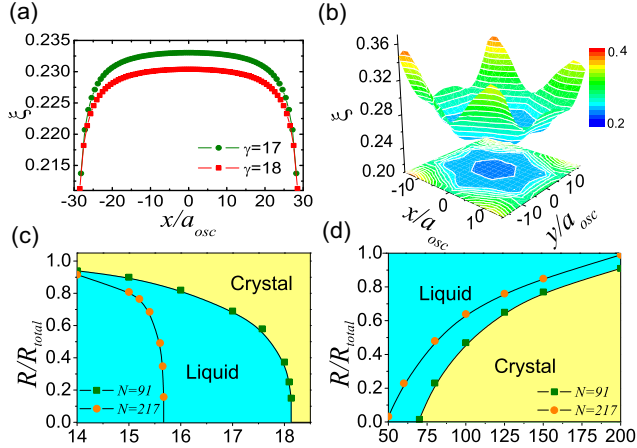


FIG. 2: (a) and (b): The distribution of softness (ξ_i) for dipolar crystals with $N = 91$ particles in 1D and 2D isotropic traps respectively. $\gamma = 80$ in (b). (c) and (d): The radii of the phase boundary, R , as a function of dipolar strength, γ , for the above two systems with $N = 91$. R_{total} is the radius of the whole system. Here we set $C_L = 0.23$, and phase boundary for $N = 217$ are also shown for comparison.

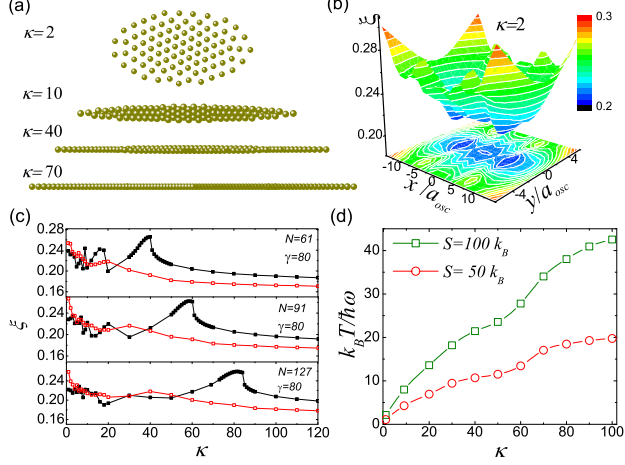


FIG. 3: (a) Position configuration of the dipolar crystal ($N = 91$) in elliptic traps of different aspect ratios, κ . (b) Distribution of the softness for $\kappa = 2$. (c) Local softness in the center (filled squares) and at the end (open squares) for systems of different numbers of particles (N). (d) The system temperature changes during an adiabatic compression from a 2D isotropic trap ($\kappa = 1$) to 1D trap ($\kappa \gg 1$) with $N = 91$. The confinement frequency in the x direction is fixed, and all results of are calculated with $\gamma = 80$.

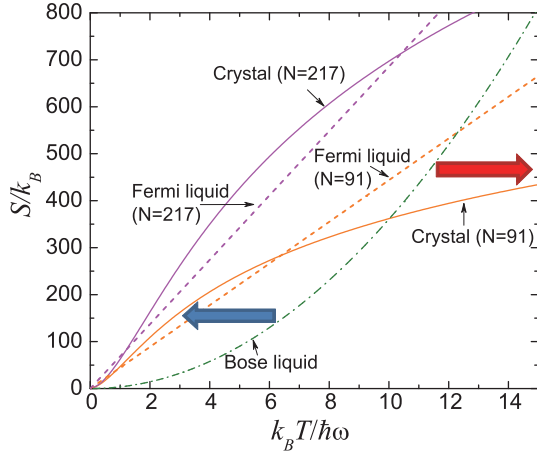


FIG. 4: Total entropy as a function of system temperature in a 2D isotropic confinement potential. Results for Bose liquid (dash-dotted lines), Fermi liquid (dotted lines) and crystal phase (solid lines) are shown together. Results for different numbers of particles are also shown for comparison. The blue leftward arrow indicates the intrinsic cooling process for a Bose liquid, while the red rightward arrow indicates the intrinsic heating process.